

Production and relevance of cosmogenic radionuclides in NaI(Tl) crystals

J. Amaré, S. Cebrián, C. Cuesta,¹ E. García, C. Ginestra, M. Martínez,² M. A. Oliván, Y. Ortigoza, A. Ortiz de Solórzano, C. Pobes,³ J. Puimedón, M. L. Sarsa, J. A. Villar, and P. Villar

Laboratorio de Física Nuclear y Astropartículas, Universidad de Zaragoza, Calle Pedro Cerbuna 12, 50009 Zaragoza, Spain

Laboratorio Subterráneo de Canfranc, Paseo de los Ayerbe s/n, 22880 Canfranc Estación, Huesca, Spain

Abstract. The cosmogenic production of long-lived radioactive isotopes in materials is an hazard for experiments demanding ultra-low background conditions. Although NaI(Tl) scintillators have been used in this context for a long time, very few activation data were available. We present results from two 12.5 kg NaI(Tl) detectors, developed within the ANAIS project and installed at the Canfranc Underground Laboratory. The prompt data taking starting made possible a reliable quantification of production of some I, Te and Na isotopes with half-lives larger than ten days. Initial activities underground were measured and then production rates at sea level were estimated following the history of detectors; a comparison of these rates with calculations using typical cosmic neutron flux at sea level and a selected description of excitation functions was also carried out. After including the contribution from the identified cosmogenic products in the detector background model, we found that the presence of ^3H in the crystal bulk would help to fit much better our background model and experimental data. We have analyzed the cosmogenic production of ^3H in NaI, and although precise quantification has not been attempted, we can conclude that it could imply a very relevant contribution to the total background below 15 keV in NaI detectors.

Keywords: dark matter, NaI detectors, radionuclide production

PACS: 95.35.+d, 29.40.Mc, 25.40.-h

INTRODUCTION

The ANAIS (Annual Modulation with NaI(Tl) Scintillators) experiment [1] aims at the confirmation of the DAMA/LIBRA annual modulation positive signal, using the same target and technique at the Canfranc Underground Laboratory (LSC). Following work on previous prototypes [2, 3], the ANAIS-25 set-up [4] consisted of two 12.5 kg NaI(Tl) crystals built by Alpha Spectra; it was intended to understand and quantify the background components and to assess the performance of the detectors. Each module consists of a cylindrical NaI(Tl) crystal, grown with selected ultrapure NaI powder and encapsulated in OFHC copper. The coupling to Hamamatsu photomultiplier tubes was performed in the clean room of LSC. ANAIS-25 was operated inside a shielding consisting of 10 cm archaeological lead plus 20 cm low activity lead.

Detectors were built in Colorado and shipped to Spain; they arrived to Canfranc on 27th November 2012 and were immediately stored underground. Data taking started just three days afterwards, allowing, together with the low background conditions and the very good energy resolution of the detectors, a precise study of the isotopes induced in the crystals by the exposure to cosmic rays; details of this work can be found at [5] and are summarized here. Data analyzed investigating the cosmogenic activation correspond to different data sets. Data sets I and II, acquired in slightly different gain conditions, span altogether for 210 days from the beginning of the data taking to the end of June 2013. Set III includes 88 days of data from March to June 2014, once the contribution of most of the cosmogenic nuclei was significantly reduced, allowing to identify longer-living products. Possible presence of tritium was evidenced in data taken from June 2014 to March 2015 and will be also discussed.

¹ Present address: Department of Physics, Center for Experimental Nuclear Physics and Astrophysics, University of Washington, Seattle, WA, USA

² Present address: Università di Roma La Sapienza, Piazzale Aldo Moro 5, 00185 Roma, Italy

³ Present address: Instituto de Ciencia de Materiales de Aragón, Universidad de Zaragoza-CSIC, Calle Pedro Cerbuna 12, 50009 Zaragoza, Spain

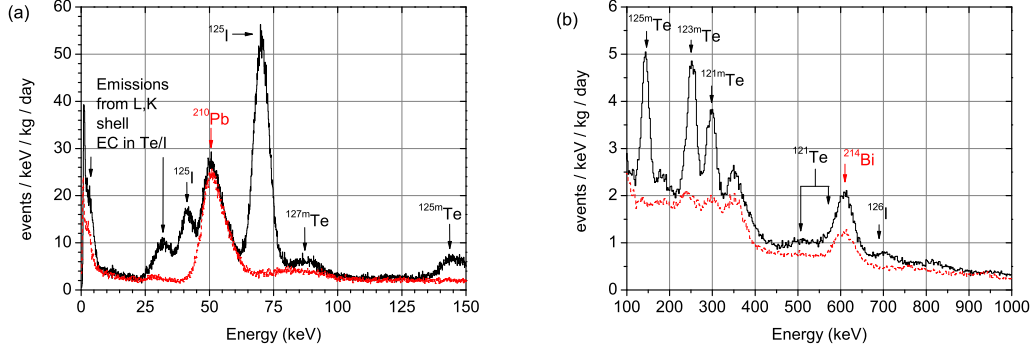


FIGURE 1. Comparison of the ANAIS-25 spectra evaluated in the beginning of data set I (black solid line) and about fifteen months afterwards, during data set III (red dashed line). The low (a) and high (b) energy regions are shown. Several cosmogenic emissions have been identified and are labeled (in black) together with the main background lines (in red).

TABLE 1. Results derived for each cosmogenic product: initial activities underground A_0 , production rates R_p obtained experimentally and calculated using different selections of the excitation function. Last column presents the ratio between the calculated and the experimental rates.

Isotope	$T_{1/2}$ (days)	A_0 ($\text{kg}^{-1}\text{d}^{-1}$)	Excitation function	Cal R_p ($\text{kg}^{-1}\text{d}^{-1}$)	Exp R_p ($\text{kg}^{-1}\text{d}^{-1}$)	Cal/Exp
^{126}I	12.93 ± 0.05	430 ± 37	MENDL-2+YIELDX	297.0	283 ± 36	1.1
^{125}I	59.407 ± 0.009	621.8 ± 1.6	TENDL-2013+HEAD-2009	242.3	220 ± 10	1.1
^{127m}Te	107 ± 4	32.1 ± 0.8	TENDL-2013+extrapolation	7.1	10.2 ± 0.4	0.7
^{125m}Te	57.40 ± 0.15	79.1 ± 0.8	TENDL-2013+HEAD-2009	41.9	28.2 ± 1.3	1.5
^{123m}Te	119.3 ± 0.1	100.8 ± 0.8	TENDL-2013+HEAD-2009	33.2	31.6 ± 1.1	1.1
^{121m}Te	154 ± 7	76.9 ± 0.8	TENDL-2013+HEAD-2009	23.8	23.5 ± 0.8	1.0
^{121}Te	19.16 ± 0.05	110 ± 12	TENDL-2013+YIELDX	8.4	9.9 ± 3.7	0.8
^{22}Na	$(2.6029 \pm 0.0008) \text{ y}$	159.7 ± 4.9	TENDL-2013+YIELDX	53.6	45.1 ± 1.9	1.2

IODINE, TELLURIUM AND SODIUM ISOTOPES

Figure 1 compares the spectra of one of the ANAIS-25 detectors evaluated in the beginning of data set I and about fifteen months afterwards, during data set III, over 13.0 and 18.5 days, respectively. Several lines can be clearly attributed to cosmogenic activation and signatures from ^{125}I , ^{127m}Te , ^{125m}Te , ^{123m}Te and ^{121m}Te were directly identified in energy spectra. ^{126}I and ^{121}Te signals were evaluated by analyzing coincidences between detectors. Although not directly seen in figure 1, ^{22}Na , having a longer mean life than I and Te products, was also identified and quantified using coincidence spectra along data set III. The cosmogenically induced activity, which is the initial activity A_0 corresponding to the moment of storing crystals deep underground at LSC, was deduced studying the exponential decay of the identifying signature produced by each I and Te isotope considering data sets I and II. For ^{121}Te , production from ^{121m}Te decay was properly taken into account. Concerning ^{22}Na , the integrated signal along data set III was used. Production rates R_p at sea level were estimated from measured A_0 values, considering that saturation was reached at detector production in Colorado (where the cosmic neutron flux is 3.6 times higher than at sea level) and a further exposition of 30 days for the boat trip from US to Spain [5]. Table 1 summarizes the results obtained for A_0 and R_p for all the identified cosmogenic products.

Production rates were also calculated considering the cosmic neutron spectrum at sea level from [6] and selecting carefully the excitation functions to minimize deviation factors between available measurements and results from different libraries and calculations from semiempirical formulae [5]. Table 1 shows also these computed production rates. Remarkable agreement with experimental rates has been achieved, specially for the products having excitation functions well validated against measurements.

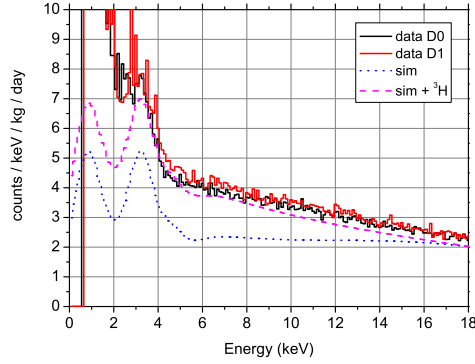


FIGURE 2. The very low energy region of the energy spectrum measured in ANAIS-25 detectors (solid lines) compared with the simulated one considering all the known and quantified intrinsic and cosmogenic activities in the detectors and main components of the set-up (dotted line) and adding also an arbitrary activity of ^3H in the NaI crystal (dashed line).

TABLE 2. Production rates of ^3H in NaI isotopes obtained considering different excitation functions.

Isotope	Library	Energy range	Production rate ($\text{kg}^{-1}\text{d}^{-1}$)
^{23}Na	TENDL-2013 [11]	<150 MeV	14.3
^{127}I	TENDL-2013 [11]	<150 MeV	15.4
^{127}I	HEAD-2009 [12]	150-1000 MeV	10.9

THE CASE OF TRITIUM

The contribution of the cosmogenic products to the background of the ANAIS-25 detectors was evaluated by Geant4 simulation, considering the measured activity values A_0 [5]. Only ^{22}Na , producing a peak around 0.9 keV, is relevant for dark matter searches. This contribution was included in the overall background model of the detectors, together with that of the known activities of the crystal and other components of the set-up (see details at [7]). When the simulated spectrum including all well-known contributions is compared with the one measured, a very good agreement was obtained except for the very low energy region. But this problem can be solved if an activity of ~ 0.2 mBq/kg of ^3H homogeneously distributed in the NaI crystal is added to the model, as shown in figure 2.

This required activity is about twice the upper limit set for DAMA/LIBRA crystals [8] and lower than the saturation activity predicted assuming the production rate of ^3H in NaI(Tl) of $31.1 \text{ kg}^{-1}\text{d}^{-1}$ calculated in [9] using production cross section estimated with TALYS 1.0 code. An attempt has been made to quantify this production rate in ^{23}Na and ^{127}I (both having 100% natural isotopic abundance in NaI) using the same approach followed for the cosmogenic isotopes identified in ANAIS-25. First, available information on the excitation function by nucleons was collected, as shown in figure 3: only one experimental result was found in the EXFOR database [10] and cross sections were taken from the TENDL-2013 (TALYS-based Evaluated Nuclear Data Library) library [11] up to 200 MeV and from the HEAD-2009 (High Energy Activation Data) library [12] from 150 to 1000 MeV. Then, the production rate was computed convoluting a selected excitation function with the energy spectrum of cosmic neutrons at sea level, using the parametrization from [6]. Table 2 summarizes the rates for several descriptions of the cross sections. The total rate considering data from TENDL-2013 library below 150 MeV reproduces the value obtained in [9], since the library is also based on TALYS code. Although information on the excitation function is very limited, it seems that the contribution to the production rate from energies above 150 MeV is not negligible. Assuming that in this high energy range neutron and proton cross sections are comparable and that production from ^{23}Na and ^{127}I is similar (as for energies below 150 MeV, see table 2) the production rate could be of $\sim 50 \text{ kg}^{-1}\text{d}^{-1}$ summing the contributions in table 2. For such a rate, an exposure of 1.9 y to the neutron flux at Grand Junction, Colorado, would produce the required ^3H activity in ANAIS-25 crystals.

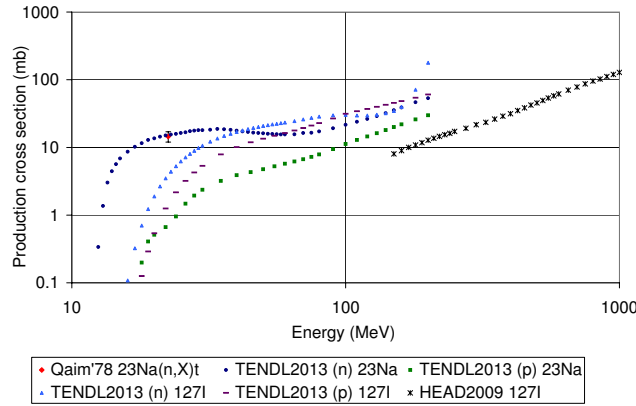


FIGURE 3. Comparison of excitation functions for production of ^3H on ^{23}Na and ^{127}I by nucleons (neutrons are indicated as n and protons as p) taken from different sources (see text).

SUMMARY

Cosmogenic activation of NaI(Tl) crystals was evaluated from measurements taken at the Canfranc Underground Laboratory using two 12.5 kg detectors produced by Alpha Spectra in Colorado (US) and shipped to Spain [5]. Production rates at sea level of a few tens of nuclei per kg and day for Te isotopes and ^{22}Na and of a few hundreds for I isotopes were derived from measured activities, in good agreement with calculations. In addition, presence of ^3H seems plausible. I and Te isotopes are not relevant after short cooling underground, but ^{22}Na and ^3H , due to their longer mean lives and low energy emissions, affect the region of interest for a dark matter search. For ANAIS-25 detectors, their contribution from 1 to 10 keV is estimated to be of 1.4 and 13.2 events/kg/d, respectively.

ACKNOWLEDGMENTS

This work was supported by the Spanish Ministerio de Economía y Competitividad and the European Regional Development Fund (MINECO-FEDER) (FPA2011-23749), the Consolider-Ingenio 2010 Programme under grants MULTIDARK CSD2009-00064 and CPAN CSD2007-00042, and the Gobierno de Aragón (Group in Nuclear and Astroparticle Physics, ARAID Foundation and C. Cuesta predoctoral grant). C. Ginestra and P. Villar are supported by the MINECO Subprograma de Formación de Personal Investigador. We also acknowledge LSC and GIFNA staff for their support.

REFERENCES

1. J. Amaré et al., *Physics Procedia* **61**, 157–162 (2015).
2. S. Cebrián et al., *Astropart. Phys.* **37**, 60–69 (2012).
3. C. Cuesta et al., *Eur. Phys. J. C* **74**, 3150 (2014).
4. J. Amaré et al., *Nucl. Instrum. Meth. A* **742**, 187–190 (2014).
5. J. Amaré et al., *JCAP* **02**, 046 (2015).
6. M. S. Gordon et al., *IEEE Trans. Nucl. Sci.* **51**, 3427–3434 (2004).
7. C. Cuesta et al., “Background analysis and status of the ANAIS dark matter project”, in these proceedings.
8. R. Bernabei et al., *Nucl. Instrum. Meth. A* **592**, 297 (2008).
9. D. M. Mei et al., *Astropart. Phys.* **31**, 417–420 (2009).
10. Experimental Nuclear Reaction Data (EXFOR) <http://www.nndc.bnl.gov/exfor/exfor.htm>, <http://www.nds.iaea.org/exfor/exfor.htm>.
11. A. J. Koning and D. Rochman, *Nuclear Data Sheets* **113**, 2841 (2012), <http://www.talys.eu/tendl-2013.html>
12. Y. A. Korovin et al., *Nucl. Instrum. Meth. A* **624**, 20–26 (2010).